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Triply resonant enhancement of third-order nonlinear optical susceptibility in compositionally asymmetric coupled quantum wells

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The third-order nonlinear optical susceptibility $\chi^{(3)}(3\omega)$ due to the intersubband transitions in the four-level AlInAs/GaInAs compositionally asymmetric coupled quantum well (CACQW) is investigated theoretically. The subband eigenenergy E_n of the CACQW structure could be designed to form an equally spaced energy-level ladder. Since the eigenenergy spacing could be designed to resonate with the pumping source, the third-order nonlinear optical susceptibility could be greatly enhanced through the triple resonance. Based on the theoretical calculations, a magnitude of $|\chi^{(3)}(3\omega)|$ as high as 2.2×10^5 (nm/V)^2 can be achieved for the CACQW structure. This is a more than eight orders of magnitude enhancement as compared to that of the bulk value in GaAs. In addition to the design of CACQW structure, the triple resonance can also be achieved by biasing the CACQW under a proper electric field due to the large Stark effect of the CACQW structure.

I. INTRODUCTION

Quantum confinement of carriers in a semiconductor quantum well[1] leads to the formation of discrete subbands. Transition between these subbands of the quantum well has extremely large oscillator strength and relatively narrow linewidth. The linear intersubband optical absorption of the quantum well has been studied experimentally and a very large and sharp optical-absorption resonance were observed.[2–6] This large dipole moment for the intersubband transition suggests that a very large optical nonlinearity may exist for the semiconductor quantum well. Recently, the optical nonlinearities associated with intersubband transitions at $\lambda\simeq10$ μm in various quantum-well structures have been theoretically investigated[7–11] and very strong second-order nonlinear effects have been observed in the multiple quantum wells under the applied electric field,[12] the step quantum well,[13–16] and the asymmetric coupled quantum wells (ACQW).[17,18] These observed second-harmonic susceptibilities are several orders of magnitude larger than that of the bulk semiconductor. Similarly, a very large third-order nonlinear optical susceptibility $\chi^{(3)}(3\omega)$ can be expected in a quantum-well structure due to its fourth power dependence on the dipole moment.[19,20] The observed third-order intersubband nonlinearity $|\chi^{(3)}(3\omega)|$ at $\lambda\simeq10.7$ μm pump wavelength measured by Sirtori *et al.*[20] in an AlInAs/GaInAs coupled-quantum-well structure is about seven orders of magnitude larger than that of the bulk value in GaAs.

The four-level AlInAs/GaInAs compositionally asymmetric coupled quantum well (CACQW) (Fig. 1) is studied here. The subband eigenenergy E_n of the CACQW structure could be designed to form an equally spaced energy-level ladder. Thus, the third-order nonlinear optical susceptibility of the CACQW can be greatly enhanced through the triple resonance. A CACQW consists of a pair of quantum wells with different depth ΔU separated by a thin barrier. The depth of each quantum well can be controlled by the composition of the quantum well. In this study, a $Ga_{0.47}In_{0.53}As$ layer is assumed for the deep quantum well, a $Ga_xIn_{1-x}As$ is assumed for the shallow quantum well, and an $Al_{0.48}In_{0.52}As$ is used as the barrier. The CACQW with this extra design parameter of the ΔU will render easy control of the subband eigenenergy level and the dipole moment. Both the eigenenergy of the ground state E_1 and the eigenenergy of the second excited state E_3 are nearly independent of ΔU, while the eigenenergy of the first excited state E_2 and the eigenenergy of the third excited state E_4 are raised with ΔU. Thus, it is possible to obtain a very large value of the third-order nonlinear optical susceptibility through triple resonance by placing E_2 in the middle of E_1 and E_3, and E_3 in the middle of E_2 and E_4. In addition, the triple resonance can also be achieved by applying a proper electric field due to the enhanced Stark effect for the CACQW structure.[21] In this article, the AlInAs/GaInAs material system is assumed for this quantum-well structure and only the left-hand-side wells are assumed to be doped with silicon (doping concentration is about 5.0×10^{17} cm^{-3}). The pumping source is assumed to be the $\lambda\simeq10.6$ μm CO_2 laser line corresponding a photo energy $\hbar\omega$ of about 117 meV.

In this article, the dependence of the third-order nonlinear optical susceptibility on the quantum-well width and the composition of the $Ga_xIn_{1-x}As$ quantum well is investigated. Subband envelope wave functions and eigenenergies are calculated self-consistently by simultaneously solving the

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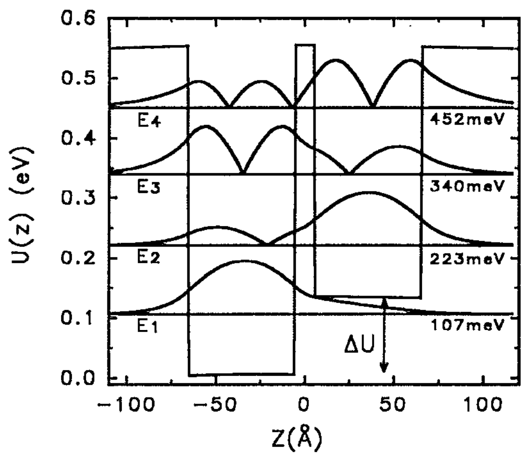

FIG. 1. Schematic diagram of an AlInAs/GaInAs CACQW structure. Both quantum wells have identical well widths of 60 Å. The barrier thickness is 11 Å and the barrier height is 550 meV. The quantum-well depth difference ΔU is 130 meV. The positions of the calculated eigenenergy subbands and the corresponding modulus square of the envelope wave function are also shown.

Schrödinger equation and Poisson's equation.[21] The Schrödinger equation is solved by the transfer-matrix method and Poisson's equation by the numerical integration. The energy-dependent effective mass due to energy-band nonparabolicity is also taking into account in this article.[22] This analysis is then employed to determine the energy-band nonparabolicity-induced lowering of the subband eigenenergy. Based on the theoretical calculation, the CACQW do give a very strong third-order nonlinear optical effect. A third-order nonlinear optical susceptibility as high as 2.2×10^5 $(\mathrm{nm/V})^2$ can be achieved for the CACQW structure. This is a more than eight orders of magnitude enhancement as compared to that of the bulk value in GaAs.

This article is organized as follows. In Sec. II a theoretical basis for the calculation is laid, and In Sec. III graphs from numerical calculations are presented along with the discussion. A conclusion is presented in Sec. IV.

II. THEORY AND FORMALISM

From the density matrix formalism, the analytic formulas of the third-order nonlinear optical susceptibility can be expressed as[23,24]

$$\begin{aligned}\chi^{(3)}(3\omega)=\frac{Nq^4}{\hbar^3}\sum_{nmvl}\Bigg(&(\rho_{mm}^{(0)}-\rho_{ll}^{(0)})\frac{M_{mn}M_{nv}M_{vl}M_{lm}}{(W_{nm}-3\omega-i\Gamma_{nm})(W_{vm}-2\omega-i\Gamma_{vm})(W_{lm}-\omega-i\Gamma_{lm})}\\&-(\rho_{ll}^{(0)}-\rho_{vv}^{(0)})\frac{M_{mn}M_{nv}M_{lm}M_{vl}}{(W_{nm}-3\omega-i\Gamma_{nm})(W_{vm}-2\omega-i\Gamma_{vm})(W_{vl}-\omega-i\Gamma_{vl})}\\&-(\rho_{vv}^{(0)}-\rho_{ll}^{(0)})\frac{M_{mn}M_{vm}M_{nl}M_{lv}}{(W_{nm}-3\omega-i\Gamma_{nm})(W_{nv}-2\omega-i\Gamma_{nv})(W_{lv}-\omega-i\Gamma_{lv})}\\&+(\rho_{ll}^{(0)}-\rho_{nn}^{(0)})\frac{M_{mn}M_{vm}M_{lv}M_{nl}}{(W_{nm}-3\omega-i\Gamma_{nm})(W_{nv}-2\omega-i\Gamma_{nv})(W_{nl}-\omega-i\Gamma_{nl})}\Bigg),\end{aligned} \tag{1}$$

where the dipole moment matrix element M_{nm} can be calculated from the following expression:

$$M_{nm}=\int_{-L_t/2}^{L_t/2}\psi_n^*(z)z\psi_m(z)dz. \tag{2}$$

L_t is the total width of the coupled quantum well, q is the charge of electron, and N is the doping concentration of the coupled quantum well. The Γ operator is the matrix composed of elements Γ_{nm} with $\Gamma_{nm}^{-1}=\tau_{nm}$ being the dephasing time between the state $|\psi_n\rangle$ and the state $|\psi_m\rangle$ and $\Gamma_{nn}^{-1}=\tau_{nn}$ being the energy relaxation time for the state $|\psi_n\rangle$. $W_{nm}\equiv\Delta E_{nm}/\hbar\equiv(E_n-E_m)/\hbar$ is the intersubband transition frequency. The diagonal density matrix element $\rho_{nn}^{(0)}$ equals to the thermal equilibrium occupation probability of the corresponding state and can be expressed as[23,24]

$$\rho_{nn}^{(0)}=\frac{1}{1+\exp[(E_n-E_F)/(K_BT)]}, \tag{3}$$

where E_F is the Fermi level of the system, T is the temperature, and K_B is the Boltzmann constant. From Eq. (1), to maximize the third-order nonlinear optical susceptibility, the relevant energy levels are equally spaced (triple resonance) and the product of the corresponding dipole matrix elements is maximum. In order to have this kind of triple resonance, eigenenergy differences $\Delta E_{21}=\Delta E_{32}=\Delta E_{43}=\hbar\omega$, $\Delta E_{31}=\Delta E_{42}=2\hbar\omega$, and $\Delta E_{41}=3\hbar\omega$ for the four-level quantum-well structures should be achieved. Thus, by properly tailoring the electronic distribution in the coupled quantum well, the third-order nonlinear optical susceptibility can be greatly enhanced. For the proposed AlInAs/GaInAs CACQW structure as shown in Fig. 1, all the relevant matrix elements are very large: $M_{12}\simeq14$ Å, $M_{13}\simeq13$ Å, $M_{14}\simeq37$ Å, $M_{23}\simeq23$ Å, $M_{24}\simeq10$ Å, and $M_{34}\simeq30$ Å. The calculated eigenenergy differences are $\Delta E_{21} \simeq 116$ meV, $\Delta E_{31} \simeq 233$ meV, and $\Delta E_{41} \simeq 345$ meV. It is evident that these eigenenergy differences observe the triply resonant condition.

To calculate the third-order nonlinear optical susceptibility, the energy eigenvalues and dipole moments for an electron in a quantum well under an applied electric field should be calculated first. For a coupled quantum well centered at $z=0$ with total well width L_t under a bias electric field normal to the well, the envelope wave function ψ_n should satisfy the following one-dimensional Schrödinger equation:

$$H\psi_n = \left(-\frac{\hbar^2}{2m^*} \frac{d^2}{dz^2} + U(z) \right) \psi_n = E_n \psi_n , \tag{4}$$

where $U(z) = -qV(z)$ represents the electronic potential energy variation, and E_n and ψ_n respectively represent the energy eigenvalue and the envelope wave function of the nth bound state. The electrostatic potential $V(z)$ in the conduction band is determined by Poisson's equation,

$$\frac{d^2V(z)}{dz^2} = -\frac{q}{\epsilon} [N_D^+(z) - n(z)], \tag{5}$$

where $N_D^+(z)$ is the ionized donor concentration and $n(z)$ is the electron density. To solve the Schrödinger equation and Poisson's equation self-consistently, an initial potential profile is first guessed. The Schrödinger equation was solved by the transfer-matrix method to find eigenenergies and envelope wave functions for the given potential profile. Poisson's equation was then solved by numerical integration to find the new potential profile for the known two-dimensional electron gas (2DEG) profile from the Schrödinger equation. This process is repeated until the convergence criterion $|V_{i+1}(z) - V_i(z)|/|V_{i+1}| < \delta$ is reached, where $V_i(z)$ is the trial potential profile, $V_{i+1}(z)$ is the resulting potential profile, and δ is a small number. The energy-dependent effective mass due to energy-band nonparabolicity is also taken into account[22] in this article. This causes a lowering of subband energies of higher excited-state subbands and the lowering effect becomes substantial for the highest-excited-state subband. For a more detailed derivation, most materials can be found in Refs. 21 and 22.

III. RESULTS AND DISCUSSION

Based on the theory developed in the previous section, the third-order nonlinear optical susceptibilities $\chi^{(3)}(3\omega)$ for the CACQWs are evaluated in this section. All the numerical calculations done in this article are based on the following parameters unless otherwise stated: the pumping source is 10.6 μm CO_2 laser line, $T=77$ K, the central barrier width is 11 Å, and all of the dephasing time τ_{nm} are assumed to have the same value of 0.14 ps. The conduction-band-gap discontinuity and effective mass m^* of the AlInAs/GaInAs material system used here is adopted from Ref. 25. The enhancement of the third-order nonlinear optical susceptibility by triple resonance is studied for various quantum-well geometry parameters such as the well width and the well depth. The electric-field dependence of the third-order nonlinear optical susceptibility is also been evaluated and shows a interesting phenomena of the field-induced triply resonant enhancement of the modulus $|\chi^{(3)}(3\omega)|$ of the third-order nonlinear optical susceptibility $\chi^{(3)}(3\omega)$.

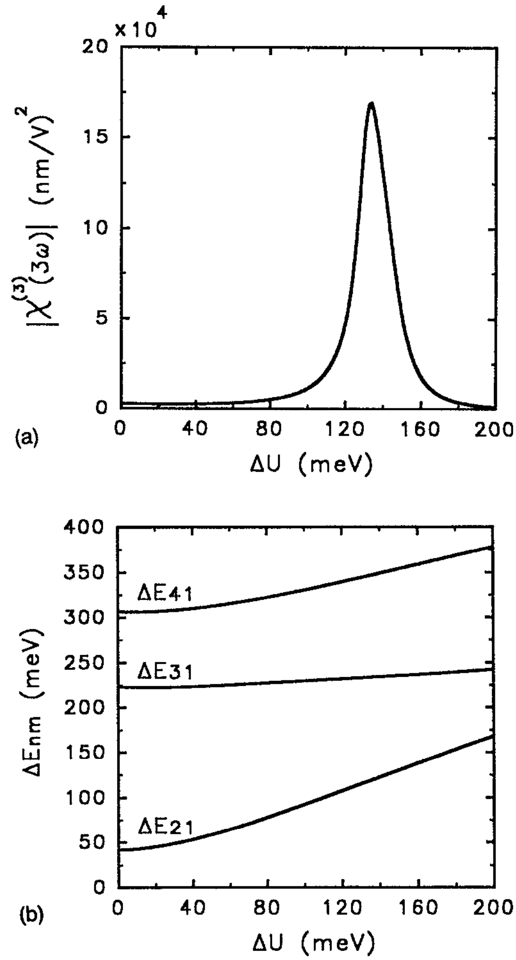

FIG. 2. (a) Calculated third-order nonlinear optical susceptibility $|\chi^{(3)}(3\omega)|$ as a function of the ΔU for the 60 Å/11 Å/60 Å CACQW. (b) Calculated eigenenergy differences ΔE_{21}, ΔE_{31}, and ΔE_{41} as a function of the ΔU for the 60 Å/11 Å/60 Å CACQW.

A. Design of the quantum-well geometry

The relation between the $|\chi^{(3)}(3\omega)|$ and the quantum-well depth difference ΔU is studied in order to optimize the $|\chi^{(3)}(3\omega)|$ for the 10.6 μm pumping source and results are plotted in Fig. 2(a). Eigenenergies of both the ground-state subband E_1 and the second excited-state subband E_3 are mostly controlled by the deep quantum well, while the eigenenergies of the first excited-state subband E_2 and the third excited-subband E_4 is mostly controlled by the shallow quantum well. As a result, the eigenenergy differences ΔE_{21} and ΔE_{41} will be increased with the ΔU and the ΔE_{31} is nearly independent of ΔU [as shown in Fig. 2(b)]. In this way, to maximize the $|\chi^{(3)}(3\omega)|$ of the CACQW through the triple resonance for the pumping photoenergy $\hbar\omega$, a coupled quantum-well structure with $\Delta E_{31} \simeq 2\hbar\omega$, $\Delta E_{41} < 3\hbar\omega$, and $\Delta E_{21} < \hbar\omega$ is designed first. Then the triple-resonant condition $\Delta E_{21} = \hbar\omega$ and $\Delta E_{41} = 3\hbar\omega$ can be obtained by increasing ΔU to the proper level. From Fig. 2(a), it is clear that the

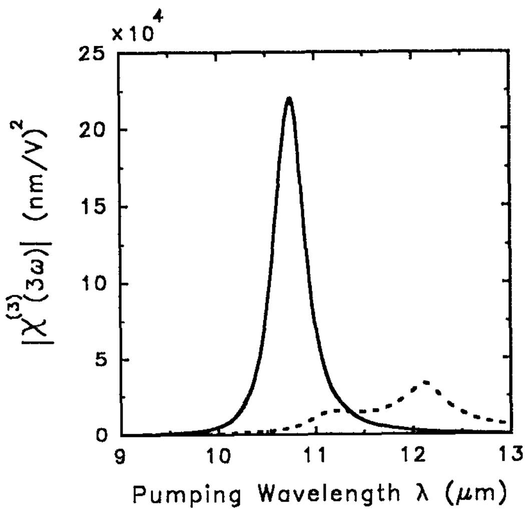

FIG. 3. Calculated third-order nonlinear optical susceptibility $|\chi^{(3)}(3\omega)|$ as a function of the pumping source wavelength for the 60 Å/11 Å/60 Å CACQW with $\Delta U=130$ meV (solid line) and the 60 Å/11 Å/60 Å CACQW with $\Delta U=0$ meV (dashed line).

$|\chi^{(3)}(3\omega)|$ peaks at $\Delta U \simeq 130$ meV which gives the triple-resonance condition of $\Delta E_{21}=\hbar\omega$ and $\Delta E_{41}=3\hbar\omega$.

In order to demonstrate the effect of the triple resonance, the $|\chi^{(3)}(3\omega)|$ as a function of the pumping source wavelength λ for the triple-resonant structure is plotted along with the $|\chi^{(3)}(3\omega)|$ of the single-resonant structure in Fig. 3. The triple-resonant structure employed here is a 60 Å/11 Å/60 Å CACQW with $\Delta U=130$ meV. As for the single-resonant structure, the 60 Å/11 Å/60 Å CACQW with $\Delta U=0$ meV is used. For the $\Delta U=0$ meV CACQW structure, $\Delta E_{21}\simeq 42$ meV, $\Delta E_{31}\simeq 223$ meV, and $\Delta E_{41}\simeq 306$ meV, the corresponding resonant peaks of the $|\chi^{(3)}(3\omega)|$ located at $\lambda \simeq 11.2$ and 12.2 μm are observed in this figure. Because of $\Delta E_{31}\neq 2\Delta E_{21}$ and $\Delta E_{41}\neq 3\Delta E_{21}$, this is a single-resonant process. As a result, both peaks of the $|\chi^{(3)}(3\omega)|$ for the $\Delta U=0$ meV CACQW structure are less than one-seventh of the peak value of $|\chi^{(3)}(3\omega)|$ of the $\Delta U=130$ meV CACQW structure. Since $\Delta E_{21}\simeq 116$ meV, $\Delta E_{31}\simeq 233$ meV, and $\Delta E_{41}\simeq 345$ meV for the $\Delta U=130$ CACQW structure, this is a triple-resonant structure. As a result, only one peak of the $|\chi^{(3)}(3\omega)|$ located at $\lambda\simeq 10.7$ μm is observed in Fig. 3. A third-order nonlinear optical susceptibility as high as 2.2×10^5 (nm/V)^2 is achieved for the 60 Å/11 Å/60 Å CACQW structure with $\Delta U=130$ meV at $\lambda \simeq 10.7$ μm. This is a more than eight orders of magnitude enhancement as compared to the bulk value in GaAs.

B. Effect of the applied electric field

The method to achieve the triple-resonance condition is not limited to tailoring the CACQW structure during the epitaxial growth. It can also be achieved through an external bias electric field. Third-order nonlinear optical susceptibilities $|\chi^{(3)}(3\omega)|$ are studied for the CACQW under an external electric field. The direction of the applied electric field is defined as from left- to right-hand side (i.e., positive z direction). The $|\chi^{(3)}(3\omega)|$ as a function of the applied electric field for the 60 Å/11 Å/60 Å CACQW with $\Delta U=100$ meV is plotted in Fig. 4(a). It is evident that the $|\chi^{(3)}(3\omega)|$ can be controlled by the applied electric field. This field dependence of the $|\chi^{(3)}(3\omega)|$ arose from the variation of dipole matrix elements and the eigenenergy differences under the bias field. The CACQW structure possesses an enhanced quantum-confined Stark effect, and a large variation of ΔE_{21} and ΔE_{41} for the CACQW structure under the applied electric field is expected [as shown in Fig. 4(b)]. Since the envelope wave function of both the ground-state subband E_1 and the second excited-state subband E_3 are symmetric, both subband levels will be lowered by the applied electric field and ΔE_{31} will stay more or less unchanged. If the CACQW is applied with a proper positive electric field, the $\Delta E_{21}=\hbar\omega$ and $\Delta E_{41}=3\hbar\omega$ conditions can be reached and the $|\chi^{(3)}(3\omega)|$ will be maximized due to the triple resonance. In this way the $|\chi^{(3)}(3\omega)|$ can be enhanced by triple resonance through the applied electric field instead of the CACQW structure.

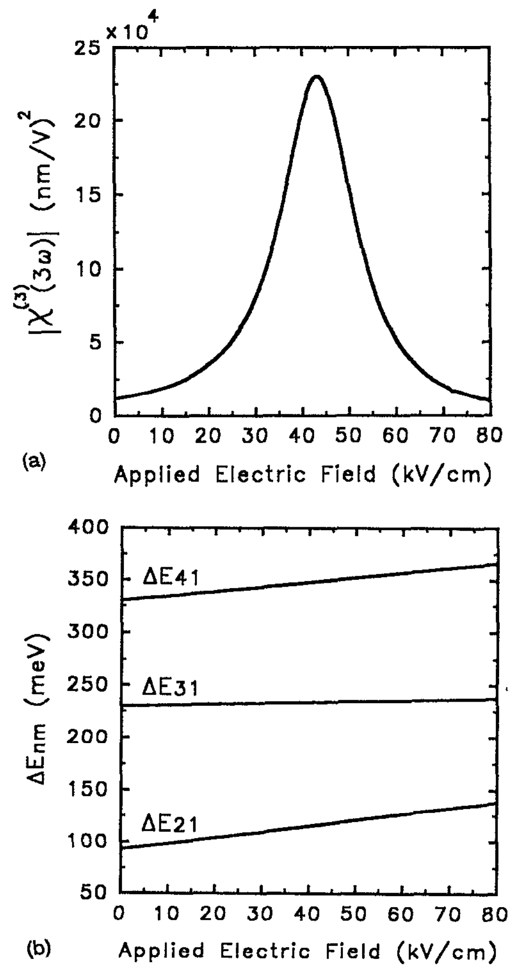

FIG. 4. (a) Calculated third-order nonlinear optical susceptibility $|\chi^{(3)}(3\omega)|$ as a function of the applied electric field for the 60 Å/11 Å/60 Å CACQW with $\Delta U=100$ meV. (b) Calculated eigenenergy differences ΔE_{21}, ΔE_{31}, and ΔE_{41}, as a function of the applied electric field for the 60 Å/11 Å/60 Å CACQW with $\Delta U=100$ meV.

IV. CONCLUSION

Several compositionally asymmetric coupled-quantum-well structures have been employed to enhance the third-order nonlinear optical susceptibility at 10.6 μm. The third-order nonlinear optical susceptibility of the CACQW has been calculated both with and without biased electric field by using the one-particle density matrix formalism. Base on the theoretical prediction, the third-order nonlinear optical susceptibility could be greatly enhanced through triple resonance. The four-level CACQW structures have been designed to yield a set of equally spaced eigenenergy levels. The third-order nonlinear optical susceptibility for this kind of CACQW structure is greatly enhanced through triple resonance. Based on the theoretical calculations, a third-order nonlinear optical susceptibility as high as 2.2×10^5 $(\mathrm{nm/V})^2$ can be achieved for the CACQW structure. In addition to the design of CACQW structure with equally spaced eigenenergy levels, the triple resonance can also be achieved by biasing the CACQW under a proper electric field due to the large Stark effect of the CACQW structure.

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